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A Survey of Selected Heavy Metal Concentrations in Wisconsin Dairy Feeds

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ABSTRACT

Heavy metals such as zinc (Zn), copper (Cu), chromium (Cr), arsenic (As), cadmium (Cd), and lead (Pb) are potential bioaccumulative toxins of the dairy production system. The heavy metal content of dairy feeds, however, remains poorly documented, particularly in the United States. This survey determined the heavy metal content of 203 typical dairy ration components sampled from 54 dairy farms in Wisconsin. Lowest heavy metal concentrations were found in homegrown alfalfa (*Medicago sativa* L.) hay and haylage, and corn (*Zea mays* L.) grain and silage. Highest metal concentrations were found in purchased feeds, particularly mineral supplements, and to a lesser extent corn- or soybean-based concentrates. Zinc and Cu were found at the highest concentration in complete dairy (total mixed and aggregated component) rations and reflected the deliberate addition of these metals to meet animal nutrient requirements although more than half the farms fed Cu and Zn above US recommended levels. Concentrations of Cr, As, Cd, and Pb were present in much lower concentrations and decreased in the order Cr > As > Pb > Cd. No complete Wisconsin dairy ration contained heavy metal concentrations above US maximum acceptable concentrations and would be unlikely to induce any toxic effects in dairy cattle. Concentrations of Cd in complete dairy rations were closest to US maximum acceptable concentrations, suggesting the greatest potential long-term risk to exceed US maximum acceptable concentrations if whole farm levels of Cd were to increase in the future. With the exception of Pb, the main sources of Zn, Cu, Cr, As, and Cd in the complete dairy feed ration originated from imported feed. The continued importation of heavy metals in dairy feed is likely to be associated with accumulation of these metals in soils where manure is applied. Although the cycling of many heavy metals through the dairy food chain will be limited by factors such as a soil's cation exchange capacity, pH, salinity, and phytotoxicity of the metal, these may be less limiting for Cd. It is important that sources of Cd in the dairy system are identified and minimized to prevent problems associated with Cd accumulation in the dairy soil system arising over the long-term.
(**Key words:** heavy metal, dairy feed, survey, Wisconsin)

Abbreviation key: MAC = maximum acceptable concentration, **RDR** = reconstructed dairy ration.

INTRODUCTION

Wisconsin has the largest number of dairy farms in the US (USDA-NASS, 2004). Traditionally, Wisconsin dairy farmers grow most of their own feed and recycle manure back to the land as a source of minerals and OM for crop production. In recent years, however, many Wisconsin dairy farms have expanded herd size without a comparative increase in land area for crop production (Bland, 2002; Jackson-Smith, 2002), resulting in greater reliance on imported feed to meet the nutritional requirements of their dairy herd and an increase in manure nutrient load applied to crops. This shift in practice, in addition to the continued nutrient inputs from other sources (e.g., agrochemicals and atmospheric deposition), has been strongly linked with a build up of soil nutrients, particularly phosphorus, at the whole-farm level (Van Horn et al., 1996; Powers and Van Horn, 2001).

Heavy metals such as zinc (Zn), copper (Cu), chromium (Cr), arsenic (As), cadmium (Cd), and lead (Pb) are potential bioaccumulative toxins of the dairy production system as soils tend to act as long term sinks for these metals (Alloway, 1995) via sorption onto metal oxides, particularly iron and manganese oxides (Brown and Parks, 2001), clay minerals, soil OM, and

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other forms of humified natural OM. Although different heavy metals display a range of different properties and mobilities in the soil, losses are generally low and may occur through crop removal, leaching, and soil erosion (Aldrich et al., 2002). The long-term accumulation of heavy metals in agricultural soils has the potential to reduce soil productivity by inhibiting soil microbial (Birch et al., 1995) and fauna (Georgieva et al., 2002) populations, and may pose a risk to animal, human, and ecosystem health (Alloway, 1995).

In a closed system, the cycling of many heavy metals through the dairy food chain is likely to be limited by the soil-plant barrier (Chaney, 1990). The soil barrier limits transmission of metals through the food chain by chemical processes that limit bioavailability. Principally this is the soil cation exchange capacity, but it is affected, in turn, by other soil chemical properties such as pH, salinity, micro- and macronutrients, and metal concentration (Alloway, 1995). Where soils receive amendments, such as biosolids or manures, bioavailability will be affected by these respective properties in the amendment (Chaney, 1990). The plant barrier limits transmission of certain heavy metals through the food chain where metals are phytotoxic, and substantial yield reduction occurs before the crop would comprise risk during lifetime ingestion by livestock. The exception to this rule is Cd. Cadmium has the greatest potential for transmission through the food chain at levels that present risk to consumers. In the 1980s, the adult population in the United States was reported to receive about 20% of the World Health Organization's allowable daily intake of Cd from the consumption of grain and cereal products (WHO, 1993).

Heavy metals may enter the dairy production system in a variety of ways. These include atmospheric deposition, land application of inorganic fertilizers, biosolids, agrochemicals, and animal manures (Nicholson et al., 2003). However, the magnitude of these direct inputs will be determined by many indirect factors, such as farm location, and the use of imported feed and fertilizer.

Zinc and Cu are essential trace minerals required for many biological processes, particularly enzyme functions, and they have a positive influence on livestock growth and reproduction. Due to the low Zn and Cu content in some homegrown feeds compared with recommendations and varying bioavailability, supplementation of these metals is necessary for most livestock species, and they are commonly added to dairy rations as mineral supplements (NRC, 1980; European Commission, 2003a,b). When these nutrients are added above requirements, however, the dairy cow may restrict undesired accumulation of Zn and Cu in tissues by adaptation of absorption and excretion leading to an increase in the Zn and Cu content of manure (Nicholson et al., 1999; McBride and Spiers, 2001).

In addition to Zn and Cu, heavy metals such as Cr, As, Cd, and Pb are generally considered contaminants of dairy feed that are imported into the ration involuntarily, generally via phosphate-containing concentrates and supplements (McBride, 2001). Ingestion of these contaminant heavy metals is likely to increase their concentration in manure. Heavy metal application to soils in the form of manure can be taken up by feed crops, perhaps exacerbating heavy metal exposure to dairy cows over the long-term.

The trivalent form of Cr is considered essential for normal carbohydrate and lipid metabolism (NRC, 1980). However, Cr (III) is ubiquitous in nature, occurring in air, water, soil, and biological materials, and supplementation is generally not considered essential. Favorable responses to Cr supplementation have been reported in swine production but not in dairy cattle, although studies are limited (European Commission, 2003c). Although Cr (III) is relatively harmless to most animal species, ingestion of a high dose (30 to 40 mg/kg of BW/d) of Cr (VI) has induced toxicosis in dairy calves (European Commission, 2003c).

Arsenic has been shown to be an essential trace element in rodents, and some organic arsenic compounds (e.g., arsanilic acid, 4-nitrophenylarsonic acid, 3 nitro-4-hydroxyphenylarsonic acid, and their salts) have been used as feed additives for disease control and improvement of weight gain in swine and poultry in high concentrations for some time (NRC, 1980). However, no evidence exists that arsenic is an essential nutrient for dairy cattle. The toxicity of arsenic is dependent on chemical form and valency. Inorganic forms of arsenic are generally much more toxic than organic forms. Ruminants are less susceptible to As toxicosis and do not show any sign of toxicity unless the feed offered contains more than 200 to 300 mg/kg of inorganic As (NRC, 1980).

Cadmium and Pb are nonessential nutrients that are of direct concern to human and livestock health and may accumulate in the body, particularly in the kidney, liver, and to a lesser extent in the muscle. Only a limited number of instances have been reported where levels in cattle tissue exceeded maximum acceptable limits for human consumption (Schwarz et al., 1991; Koh et al., 1998), but recent work has suggested that dairy cattle may be more susceptible to the accumulation of Cd and Pb than beef cattle (Alonso et al., 2003). Although it is unlikely that Cd would accumulate in products intended for human consumption, accumulation has been observed in the ovaries and uteri of dairy cows (Smith, 1986) that may have an impact on reproduction.

Most work concerning heavy metals in animal feed has focused on pig and poultry production where it is common practice to add Cu and Zn at rates much higher than animal requirements. Very little is known about the heavy metal concentration of dairy feeds and feed components. Typical concentrations of Cu and Zn have been published in the United States (NRC, 1980, 2001) and Europe (European Commission, 2003a,b,c), but only a few studies have reported concentrations of contaminant heavy metals (e.g., Pb and Cd), which are of more concern for animal and human health (Nicholson et al., 1999; Olson et al., 2002b), and no study could be found that gives typical heavy metal concentrations for Wisconsin dairy feeds. The objective of this study was to determine heavy metal (Zn, Cu, Cr, As, Pb, and Cd) concentrations found in typical dairy rations and ration components from representative Wisconsin dairy farms.

MATERIALS AND METHODS

Farm Selection and Dairy Diet Component Classification

A stratified random sample of 54 dairy farms was drawn from a subset of 260 located in the prominent dairy counties of Wisconsin. Each farm was visited in the spring of 2003, and a structured survey was used to collect general information on farm practices and characteristics and identify different feeding groups from within the lactating and heifer herds. Selected characteristics of the sample farms are shown in Table 1. The types and amount of feed being fed on the day of the interview were recorded. Feed samples were taken from both the lactating and growing heifer (6 mo to breeding) feed group as individual components or as a TMR.

Forage samples could be classified as alfalfa (*Medicago sativa* L.) hay and haylage, and corn (*Zea mays* L.) silage. A further classification called "forage mixtures" was developed for use when individual forage components were not available. Forage mixtures were typically near equal proportions of alfalfa haylage and corn silage but may also have contained mineral supplements or concentrates. Forage mixtures however, could not be considered a TMR, as in each case additional concentrates, and in some instances forages, were fed.

Concentrate samples could generally be classified into 4 categories; corn grain, corn-based grain mixtures, soybean (*Glycine max* L.)-based protein mixtures, and mineral supplements. Corn grain was shell corn that received no other additions. Corn-based grain mixtures were defined as mixtures prepared by a feed mill containing at least 50% processed corn grain and other ingredients in varying proportions [e.g., barley grain (*Hordeum vulgare* L.), soybeans, mineral supplements]. Soybean-based protein supplements were defined as mixtures prepared by a feed mill containing at least 40% soybean meal and other ingredients in varying proportions (e.g., whole cottonseeds, distillers grain, corn grain, molasses, mineral supplements). Mineral supplements included P-containing mineral nutrients (e.g., dicalcium phosphate, monophosphate), trace mineral mixes, buffers, and limestone.

Sample Collection

All baled, loose, and chopped alfalfa hay was sampled using a stainless steel (2.9 cm diameter × 45.7cm long) bale corer (Penn State Forage Probe, Nasco Co., Fort Atkinson, WI) attached to a portable electric drill. At least 12 cores were randomly taken from different areas of the hay lot, taking care to avoid any spoiled material. In this instance, a "lot" was defined as hay being fed by the farmer at the time of sampling. Cores were taken by inserting the probe from the center of the "butt end" of each bale to approximately three-quarters of the bale length. Individual cores were bulked together and thoroughly mixed before a composite sample of approximately 500 g was placed in an airtight plastic bag, voided of air, and sealed.

Corn silage, alfalfa haylage, forage mixtures, and TMR were sampled during the feeding period. At sampling, the feed cart was initially allowed to run for approximately 90 s, and then at least 12 grab samples were taken at roughly 5-s intervals, taking care to avoid any spoiled material. Individual grab samples were bulked together and thoroughly mixed. A composite sample of approximately 1 kg was placed in an airtight plastic bag, voided of air, and sealed.

Concentrates and supplements were sampled by taking grab samples from at least 12 regions within their respective storage areas. Individual grab samples were bulked and thoroughly mixed, and then a composite sample of approximately 500 g was placed in an airtight plastic bag, voided of air, and sealed. The exceptions were corn grain and mineral mixtures where 1 kg and 50 g composite samples were taken, respectively.

All collected samples were immediately refrigerated (Koolatron Manufacturing, Chicago, IL), brought back to the laboratory and frozen (–25°C) for later heavy metal analysis.

Table 1. Comparison of selected dairy farm characteristics and management practices from survey sample pool and Wisconsin statewide average.

Farm characteristic	Sampled farms	Statewide benchmark[1]
Total number of farms	54	N/A
Lactating herd size		
Mean herd size	88	91
Median herd size	60	56
Productivity		
Milk shipped (kg/cow per d)	29	27
Milking and housing facilities (% of farms)		
Stanchion or tie stall barn	83	82
Parlor (flat barn, pit, or other parlor)	17	18
Free stall barn for milking herd	31	20
Dairy management practices (% of farms)		
Balances feed rations at least 4 times/yr	78	64
Keeps production records on individual milk cows	73	63
Uses TMR machinery	39	37
Cropland operated (ha)		
Mean	111	103
Median	80	73

[1]Buttel et al. (1999).

Heavy Metal Analyses

Heavy metal concentrations in feed samples were analyzed according to the procedures outlined by Peters (2003) using ultrapure trace metal grade reagents. Feed samples were dried to a constant mass at 60°C and ground through a 1-mm plastic screen (Wiley mill; Arthur H. Thomas, Philadelphia, PA). A 0.5-g subsample of the ground feed was transferred to an acid-washed, 50-mL folin digestion tube and carefully digested in 5 mL of concentrated nitric acid and 1 mL of 30% hydrogen peroxide solution at 120°C using a digestion block. Digestion was considered complete when production of reddish-orange fumes and foam within the tube had subsided, the solution had become clear, and did not bubble or react upon agitation. Tubes were removed from the digestion block, cooled, diluted to 50 mL using deionized water, and stored in high-density polyethylene plastic bottles (Nalgene, Sybron Corp., Rochester, NY) at 5°C until analysis.

Zinc and Cu concentrations in digestion solutions were determined by inductively coupled plasma optical emission spectrometry (Jarrell-Ash IRIS High Resolution ICP-OES, Genesis Laboratory Systems, Grand Junction, CO). Detection limits were 0.1 ppm in the digested solution. Chromium, As, Cd, and Pb concentrations in digestion solutions were determined by inductively coupled plasma mass spectrometry (VG PlasmaQuad PQ2 Turbo Plus ICP-MS, Thermo Electron Corporation, Milford, MA). Detection limits were 0.1 ppb in the digested solution.

Analytical results were verified using National Institute of Standards and Technology (NIST) standard plant tissue reference materials. A 15% paralleled replication of samples was used as a quality control procedure (Nicholson et al., 1999).

Diet Composition

The proportion of diet components constituting a typical Wisconsin dairy ration was calculated as a percentage of overall DMI according to information supplied by the producer. Where feed rations were collected as a TMR, percentage DMI of each feed component was calculated according to TMR feed sheets supplied by the producer. Where diets were calculated by the reconstruction of dairy rations, DMI was calculated using laboratory analysis in conjunction with fresh weight intake of feed components provided by the producer. Three of the original 21 farms where feed had been collected as TMR did not wish to continue in the study and were excluded. Reconstruction of complete dairy rations for 10 of the original 33 farms where feed rations had been collected as individual components were not included, either because of inaccurate feed ration information or missing samples of feed components.

Statistical Analysis

Heavy metals concentrations in feed components and complete rations were analyzed using the general descriptive statistics component of SAS (SAS Institute, 1990).

Table 2. Concentration of zinc and copper (mg/kg of DM) in typical components of Wisconsin dairy herd feed ration.

Feed description	n	Heavy metal	Mean	SD	Median	Max	Min	Literature values
Homegrown								
Alfalfa hay	43	Zn	23	8	21	54	13	28[1,2]
		Cu	6.8	2.1	6.6	11.4	3	7.3 to 9[1]
Alfalfa haylage	26	Zn	24	7	23	55	17	28[1,2]
		Cu	6.8	1.3	6.6	10.0	5	7.3 to 9[1]
Corn silage	20	Zn	27	13	24	77	17	27 to 29[1,2]
		Cu	4.0	1.2	3.8	6.4	2	7.6[5]
Corn grain	16	Zn	25	11	23	52	12	27 to 29[1,2]
		Cu	3.7	5.3	2.1	22.3	1	1.9 to 3.3[1]
Imported								
Corn grain mix[3]	29	Zn	154	122	118	495	23	
		Cu	38.2	46.7	22.2	231.5	3	
Soybean protein mix[4]	33	Zn	165	175	91	840	20	
		Cu	45.9	72.8	18.3	420.6	4	
Mineral mix[5]	21	Zn	3060	2170	2978	6980	45	
		Cu	559.9	375.5	550.5	1338.9	4	

[1]Typical values reported by the European Commission (2003a).

[2]Typical values reported by NRC (2001).

[3]Corn-based grain mixtures containing at least 50% processed corn grain.

[4]Soybean based-protein mix containing at least 40% soybean meal.

[5]Mineral mix supplements included phosphorus-containing mineral nutrients, trace mineral mixes, buffers, and limestone.

RESULTS AND DISCUSSION

Diet Composition

The farms in this study (n = 41) typically fed lactating dairy cows diets containing 61% forage and 39% (SD ± 10) concentrates. Forage constituents were always alfalfa hay, alfalfa haylage, or corn silage but proportions varied considerably. In general, forage consisted of 47% (±31.3) alfalfa haylage, 28% (±18.8) corn silage, and 25% (±30.3) alfalfa hay. Alfalfa (either haylage or hay) was fed by every producer. Alfalfa hay was fed as a sole source of forage by 2 producers, whereas alfalfa haylage was fed as a sole source of forage by one producer. Corn silage was fed on only 80% of farms and it never exceeded a maximum of 66% of the total forage intake.

Concentrates constituents varied considerably. Corn grain or corn-based grain mixes were fed on all farms except one, and typically constituted 71% (±31.3) of the total concentrates fed. Aside from the above producer, the minimum amount of corn grain or corn-based grain mixes in concentrates was 44%, and 6 instances were recorded where it was the only concentrate fed. Typically, 26% (±17.3) of fed concentrates were soybean (TMR constituents only) or soybean meal-based protein mixes, with a maximum of 88% of total fed concentrates for the producer that fed no corn grain. Soybean or soybean meal-based protein mixes were fed on all farms with the exception of the 6 farms that fed only corn grain mixes as a sole concentrate. The remaining 3% (±6.42) could be classified as other constituents. In the case of diets fed as individual components, these were only mineral mixtures, but where diets were fed as a TMR, cottonseed and distillers grain were also included.

Concentration of Heavy Metals in Feed Components

The typical concentrations of essential and contaminant heavy metals in typical components of Wisconsin dairy feed rations are presented in Tables 2 and 3, respectively. Heavy metal concentrations in forage mixes are not presented as they yielded little additional information. Heavy metal concentrations were used in the reconstruction of complete dairy rations presented in Table 4.

Zinc

Zinc was detected in all dairy feed components (Table 2). The concentration of Zn in Wisconsin alfalfa was slightly lower than typical values reported for the United States (NRC, 2001). High levels of Zn (approximately 300 mg/kg) have been found in the tissue of alfalfa grown in low pH and high Zn soils but resulted in a 20 to 90% yield reduction (Ibekwe et al., 1996). Zinc in corn silage and corn grain was within typical

Table 3. Concentration of chromium, arsenic, cadmium, and lead (μg/kg of DM) in typical components of Wisconsin dairy herd feed ration.

Feed description	n	Heavy metal	Mean	SD	Median	Max	Min	Detection (%)
Homegrown								
Alfalfa hay	43	Cr	709	572	542	3692	252	100
		As	97	89	65	310	ND	90.7
		Cd	74	85	48	467	17	100
		Pb	198	217	166	1234	ND	76.7
Alfalfa haylage	26	Cr	906	340	854	2052	513	100
		As	170	154	119	714	31	100
		Cd	74	98	55	548	24	100
		Pb	271	190	229	726	ND	92.3
Corn silage	20	Cr	519	356	389	1702	207	100
		As	201	734	23	3306	ND	70.0
		Cd	62	126	29	593	12	100
		Pb	260	664	121	3039	ND	75.0
Corn grain	16	Cr	326	276	243	1076	69	100
		As	71	146	4	469	ND	56.3
		Cd	181	159	183	433	ND	87.5
		Pb	134	232	67	920	ND	62.5
Imported								
Corn grain mix[1]	29	Cr	1829	1984	1023	9904	164	100
		As	1022	905	814	3454	ND	96.6
		Cd	342	154	375	756	43	100
		Pb	250	373	175	1921	ND	75.9
Soybean protein mix[2]	33	Cr	1648	2002	1007	11,160	118	100
		As	683	945	357	4030	20	100
		Cd	259	140	204	580	39	100
		Pb	237	489	90	2764	ND	66.7
Mineral mix[3]	21	Cr	69,349	61,872	47,092	22,6321	1563	100
		As	10,554	11,644	5722	46,916	290	100
		Cd	1581	733	1649	3206	404	100
		Pb	2857	2483	1791	9468	175	100

[1]Corn-based grain mixtures containing at least 50% processed corn grain.

[2]Soybean-based protein mix containing at least 40% soybean meal.

[3]Mineral mix supplements included phosphorus-containing mineral nutrients, trace mineral mixes, buffers, and limestone.

ranges reported in both the United States (NRC, 2001) and the European Union (European Commission, 2003a), and in independent studies from the United Kingdom (Nicholson et al., 1999) and the United States (Olson et al., 2002a). High levels of Zn (approximately 400 mg/kg) have been reported in the tissue of corn plants grown on a peat soil with high Zn content and mobility, but extreme phytotoxic effects on yield were observed (Martinez et al., 2002). Several studies have reported an increase in the Zn concentration of corn tissues in response to an increase in soil Zn concentrations (Vanderwatt et al., 1994; Jahiruddin et al., 2001) although maximum corn tissue concentrations were within the normal range. These results indicate that the occurrence of high concentrations of Zn in alfalfa or corn intended for dairy consumption is unlikely as the phytotoxicity of the element is such that yield reductions are likely to prevent their inclusion in the dairy ration.

As expected, imported grain and protein mixtures and mineral mixtures generally contained much higher levels of Zn than those observed in the homegrown feeds, and variation was higher. This reflects the deliberate addition of Zn to these rations to meet the Zn requirements in the complete dairy ration.

Copper

Copper was detected in all feed components (Table 2) but at lower concentrations than Zn. However, the trend in Cu and Zn concentrations across the different feed groups was very similar. Copper concentration in alfalfa, corn silage, and corn grain were within typical ranges reported in the United States (NRC, 2001) and the European Union (European Commission, 2003b). Results for corn silage were also similar to results from independent studies from the United Kingdom (Nicholson et al., 1999) and the United States (Olson et al., 2002a). It would seem likely that, as with Zn, the soil-plant barrier limits Cu concentrations in dairy feed. The effectiveness of the plant barrier was demonstrated by McBride and Spiers (2001) who grew corn

Table 4. Concentration of zinc, copper, chromium, arsenic, cadmium, and lead in complete dairy rations.

Ration description[1]	Heavy metal	Mean	SD	Median	Max	Min	Recommended concentration	Maximum acceptable concentration
		mg/kg of DM						
TMR-L	Zn	84	30	76	141	44	63,[2] 47,[3] 50[4]	500[7]
RDR		82	56	75	300	28		
TMR-H		66	24	67	95	27	33,[2] 25,[3] 40 to 50[4]	
TMR-L	Cu	19	8	17	43	9	9 to 11,[2] 10[4,5]	100[7]
RDR		17	8	16	35	6		
TMR-H		22	11	22	43	6	9 to 10,[2] 10[4,5]	
		μg/kg of DM						
TMR-L	Cr	1425	952	1138	4604	613	Not established	1,000,000[6,8]
RDR		1418	772	1134	3768	456		3,000,000[6,9]
TMR-H		2135	1384	1648	5429	800		
TMR-L	As	433	316	420	1217	55	Not established	50,000[6,10]
RDR		490	398	422	1633	33		1,000,000[6,11]
TMR-H		450	370	288	1236	112		2000[7]
TMR-L	Cd	51	19	46	108	22	Not established	500[6]
RDR		159	79	141	296	53		1000[7]
TMR-H		63	29	55	128	33		500[6,7]
TMR-L	Pb	97	261	ND	1122	ND	Not established	30,000[6]
RDR		259	296	199	1480	43		5000[7]
TMR-H		274	222	224	734	ND		

[1]TMR-L = TMR for lactating herd, n = 18; RDR = reconstructed dairy ration for lactating herd, n = 23; TMR-H = TMR for growing heifer herd, n = 12.

[2]United States (National Research Council, 2001).

[3]United Kingdom (Agricultural Research Council, 1980).

[4]Germany (Gesellschaft für Ernährungsphysiologie, 2001).

[5]France (Institut National de la Recherche Agronomique, 1989).

[6]United States (National Research Council, 1980).

[7]European Union (European Commission, 2003c).

[8]Chromium oxide.

[9]Chromium chloride.

[10]Inorganic arsenic.

[11]Organic arsenic.

in soils of varying Cu concentrations and found very limited Cu accumulation in plant tissue (<35 mg/kg), even under the most extreme concentrations due to Cu-induced root damage.

As with Zn, imported grain and protein mixtures and mineral mixes generally contained much higher levels of Cu concentrations, although variance was large. This reflects the deliberate addition of Cu to these rations to meet the Cu requirements in the complete dairy ration.

Chromium

Chromium was detected in all feed components (Table 3) and was generally found in higher concentrations than other contaminant heavy metals (i.e., As, Cd, and Pb). The trend in Cr concentrations across the different feed groups was similar to Cu. The lowest average and median concentrations of Cr were found in homegrown feeds, with corn being slightly lower than alfalfa. Few studies have reported typical Cr concentrations of forages. Two previous independent surveys from the United Kingdom (Nicholson et al., 1999) and the United States (Olson et al., 2002b) reported lower Cr in corn silage than the level found in this Wisconsin study.

Imported grain and protein mixtures generally contained higher levels of Cr than forages. Mineral mixtures, however, contained average and median Cr concentrations about 120 times higher than that measured in homegrown feeds, although variance was large. These results are consistent with those of Sullivan et al. (1994) who found that the Cr content of minerals used as feed ingredients were highly variable, particularly P-containing minerals, ranging from 60 to 500 mg/kg with average values around 200 mg/kg. In addition, blending of feed components in stainless steel containers and processors, which typically contain 18% Cr, may also contribute to Cr contamination and increased Cr levels in feedstuffs.

Arsenic

Arsenic detection levels were extremely variable across different feed components (Table 3). In general, As levels in homegrown forages were well below levels that could be considered toxic, although variance was extremely high, particularly for corn grain and silage due to extremely high As levels determined at 2 farms. Levels at one farm exceeded the European Union (European Commission, 2003a) maximum acceptable concentration (**MAC**) of 2 mg/kg for As in complete feedstuffs but was considerably lower than the MAC of 100 mg/kg for organic US dairy feeds (NRC, 1980). Few studies have reported typical As concentrations of forages, though comparison of median values for corn silage were within the range of levels reported by Nicholson et al. (1999) of <1 mg/kg in the United Kingdom and were consistent with observation (European Commission, 2003c) that As levels in feed rarely exceed 0.3 mg/kg. The main mechanism for As accumulation in agricultural soils is through the use of arsenical pesticides (Adriano, 2001). This practice is particularly prevalent in orchards. Elevated levels of As have been found in pastures previously used as orchards (Willett et al., 1993). Several of our study regions in Wisconsin have a long history of cider production and have many orchards. Subsequent farm visits are planned to identify the history of the farms where high levels of As were found.

Imported grain and protein mixtures generally had about 10 times higher levels of As than found in homegrown feeds, and likely reflects contamination via the addition of other components to mixes by the feed mill (e.g., P-containing mineral mixtures). The highest concentration of As was found in the mineral mixtures. Although these components generally exceed MAC, they are not intended as a complete feedstuff but as part of a formulated ration. However, as it is unlikely that such a ration is formulated based on As levels, elevated levels in forage are some concern as these generally serve to dilute the anticipated high concentration from sources containing mineral mixture. However, in this instance the high levels of As in the corn silage of the particular farm was not reflected in the overall which was below even the lower limits set by the European Union (European Commission, 2003c).

Cadmium

Cadmium was detected in all samples except corn grain, where Cd was not detected in 3 of the 16 samples (Table 3). The lowest average and median concentrations of Cd were generally found in forages but variance was large. Few studies have reported typical Cd concentrations of forages. A UK study (Nicholson et al., 1999) found average Cd in corn silage to be higher than those reported here (approximately 160 μg/kg) and other forages (e.g., grass silage) were found to have concentrations of <1000 μg/kg.

Although Cd levels were generally low in Wisconsin forages, 2 farms had forage components (corn silage and alfalfa haylage) that slightly exceeded the permissible limit of 0.5 mg/kg in US feeds (NRC, 1980), but not the European Union's 1 mg/kg limit for complete feedstuffs (European Commission, 2003c). Reasons for the elevated levels in these forages were unclear but as Cd is generally more mobile in the soil and plant than other heavy metals, this finding may reflect elevated soil Cd levels in the soils of these farms possibly through the application of biosolids or other industrial contamination. High concentrations of Cd (up to 10 mg/kg) have been found in forages grown in fields near industrial zinc-plating sites, where urban sludge has been used as a fertilizer and where silt from industrial areas has been deposited (Smith, 1986). Similarly, Miller et al. (1995) measured yield and shoot concentrations of Cd in alfalfa of up to 0.64 mg/kg grown in soils receiving high rates of sewage sludge (equivalent to 4.6 kg of Cd/ha). They also noted that although Cd concentrations in alfalfa did not reach phytotoxic levels, most treatments equaled or exceeded the 0.5 mg of Cd/kg that is permissible in US animal feed (NRC, 1980).

Higher mean and median Cd levels were found in corn grain, although variance was extremely high. It was difficult to explain these results. Although Cd is generally more mobile in the soil and plant than other heavy metals, its concentration in corn would be expected to decrease in the order: root > leaves > stem > subterranean storage organs > grain. It is possible that strains of corn grown for grain have a greater Cd uptake potential compared with strains grown for silage, but such a conclusion cannot be drawn from this data set and requires further investigation.

Imported grain and protein mixtures generally contained median Cd concentrations 4 times higher than homegrown feeds. The highest Cd concentrations were found in mineral mixtures, but only at levels comparable with those reported for other feeds, such as wheat grain (WHO, 1993).

Lead

Concentrations of Pb in dairy feeds were extremely variable and 100% detection only occurred for the mineral mixture component. Concentrations of Pb in all feeds were generally higher than Cd or As, but lower than Cr. High variation made it difficult to determine trends in concentration levels across feed types, but in general, the lowest concentrations of Pb were found in corn grain and the highest in mineral mixtures.

Concentrations of Pb for corn silage were within the range reported by Nicholson et al. (1999) of < 1 mg/kg in the United Kingdom and generally lower than values reported in Germany of approximately 2 mg/kg (KTBL, 2004). The availability of Pb in soils is generally low. In a long-term study of Pb uptake by corn on soils receiving high application levels of Pb via sewage sludge, Schaecke et al. (2002) found that plant uptake of heavy metals was <1% of the total amount applied. It seems likely that the high variation associated with Pb determinations, particularly in forages, may have been partly due to Pb from other sources, such as airborne contamination.

The concentration of Pb in imported grain and protein mixtures was generally lower than the 2 mg/kg reported in Germany (KTBL, 2004), although several samples exceeded this level. Similarly, mineral mixtures were on average lower than the 5.2 mg/kg reported for German cattle mineral mixtures (KTBL, 2004).

Concentration of Heavy Metals in Complete Dairy Feed Rations

The heavy metal concentrations in complete dairy rations are presented in Table 4. Total mixed rations were sampled as complete rations and did not include any of the individual components described in Tables 2 and 3. Reconstructed dairy rations (**RDR**) were calculated as the proportionate combination of heavy metal concentrations and DM of each feed component. Calculation of RDR was not possible for 10 of the survey farms because of inaccurate feed ration information or missing samples of feed components. Similarly, heifer diets were not reconstructed due to the difficulty in estimating DMI because forage was generally fed ad libitum.

Average Zn concentrations in total dairy rations determined in TMR samples or calculated by RDR were similar. On average, 50% of farms feeding TMR and 59% of farms feeding RDR fed Zn above US (NRC, 2001) recommended levels (Table 4). Average Zn concentrations for growing heifers were lower than those for lactating cows, but still exceeded recommended levels on 75% of the farms. The apparent overfeeding of Zn even up to the maximum determined amount (300 mg/kg) was lower than the MAC for US feeds (NRC, 1980). It is highly unlikely that any of the dairy cattle in this study would experience any signs of Zn toxicity. Indeed, dairy cows have been shown to tolerate dietary Zn concentrations of up to 1300 mg/kg (Archibald, 1944; Ott et al., 1966) and heifers have been shown to be unaffected by dietary Zn levels of 500 mg/kg, although levels of 900 mg/kg caused reduced weight gain and lowered feed use efficiency (Ott et al., 1966).

As with Zn, average concentrations of Cu determined by TMR and RDR were similar. Determined average Cu levels of all diets exceeded NRC (2001) recommended levels by, on average, 7 mg/kg (Table 4). Overall, 94% of farms' TMR and 75% of farms' RDR had Cu concentrations that exceeded NRC (2001) recommended levels. The average Cu concentrations of feed intended for growing heifers were higher than those for lactating cows, but still exceeded recommend levels for this group on 75% of farms. However, the apparent overfeeding of Cu even up to the maximum determined amount (43 mg/kg) was lower than the MAC for US feeds (NRC, 1980). It is highly unlikely that any of the dairy cattle in this study would experience any signs of Cu-induced toxicity. Generally, only calves are susceptible to Cu toxicity because Cu absorption declines considerably when the function of the reticulorumen develops. Copper toxicity in dairy calves has been shown in calves at Cu concentrations of 30 mg/kg (Agricultural Research Council, 1980).

Although feeding Zn and Cu above recommended levels does not pose any risk to the dairy cow, it may lead to an increase in the Zn and Cu content of manure (Nicholson et al., 1999; McBride, 2001) and may contribute to the long-term accumulation of these metals in the dairy soil system.

The concentration of contaminant heavy metals in the dairy ration were typically found in the following order Cr > As > Pb > Cd, although the order of Pb and Cd could change depending on a particular farm. Determined average concentrations of Cr and As in the dairy rations were not affected by the method of analysis; similar concentrations of Cr or As were found for both TMR or RDR rations. Lead and Cd concentrations, however, showed lower levels in TMR samples compared with levels determined by RDR. This finding is most likely due to the lower detection limits associated with calculated RDR method, rather than a consequence of the feed method used by the producers.

None of the complete dairy feed rations had concentrations of any contaminant heavy metal above MAC for US dairy feed (NRC, 1980). It can be concluded that it would be unlikely that any of the complete Wisconsin dairy rations would induce any toxic symptoms in dairy cattle from associated levels of these contaminant heavy metals. Using median values from RDR, our results suggest that, of all the contaminant heavy metals, Cd levels were closest to appropriate MAC for US dairy feeds (NRC, 1980) with levels 3.5× lower than current MAC compared with 150× lower than for Pb, 118× lower than for As (inorganic) and 882× lower than for Cr.

Although the concentrations of the contaminant heavy metals are below limits likely to induce symptoms of toxicity in dairy cattle, the continued importation of these minerals in dairy feed is likely to be associated with accumulation of the metals in soil where manure is applied repeatedly over the long term. The slow accumulation and limited plant bioavailability of Pb, As, and Cr are unlikely to cause buildups that may significantly increase the concentration of these elements above MAC in the dairy ration, but this may not be the case for Cd. It is important that sources of Cd in the dairy system are identified and minimized to prevent problems from arising in the long term.

Sources of Heavy Metals in Complete Dairy Feed Rations

In this study, homegrown alfalfa hay, alfalfa silage, corn silage, and corn grain are classified as feed components that contain no additional ingredients from off-farm sources. Imported feeds are defined as those that contain ingredients added by the feed mill, and include corn-based grain mixes, soybean-based protein mixes, and mineral supplements.

According to results from RDR (n = 23), producers typically fed a ration containing an average of 68% homegrown and 32% (SD ±15.02) imported ration components. Imported components contributed on average 73 and 67% of the total intake of Zn and Cu, respectively, with the remaining 27 and 33% (SD ±14) provided by homegrown components. This was expected because Zn and Cu are deliberately added to dairy ration formulations to combat deficiencies in these elements.

As with Zn and Cu, imported components of the dairy ration were major contributors toward the total intake of Cr, As, and Cd for RDR diets. On average, imported components contributed 57, 68, and 65% of the total intake of Cr, As, and Cd, respectively, with the remaining 43 (SD ± 21), 32 (SD ± 27), and 35% (SD ± 25) provided by homegrown components. This suggests that the feed industry may need to address the sources of these contaminant metals, particularly Cd. In contrast to the other heavy metals, the major contributor toward the total intake of Pb in the dairy ration was from homegrown feed, which contributed 62% of the total intake of Pb, with imported components contributing 38% (SD ± 33). This may reflect the atmospheric deposition of Pb onto growing crops from sources such as diesel engines or industry.

CONCLUSIONS

The Wisconsin dairy farms in this study typically fed lactating dairy cows diets containing 61% forage (alfalfa hay and haylage and corn silage) and 39% concentrates (corn grain, corn-based grain, and soybean based-protein mixes, and mineral mixes). The majority of the forage components had heavy metal (Zn, Cu, Cr, As, and Cd) concentrations similar to or lower than the few previous studies, suggesting that levels or availability of these metals in Wisconsin dairy system may be limited. The exception was Cd, where concentrations above the maximum acceptable concentration for US feedstuffs (NRC, 1980) were found in forages on 2 farms.

The heavy metals found at the highest concentration in complete dairy ration (TMR and aggregated component rations) were Zn and Cu, reflecting the deliberate addition of these metals to meet animal nutrient requirements. However, more than half the farms were feeding Cu and Zn above NRC (2001) recommended levels. Concentrations of Cr, As, Cd, and Pb were present in much lower concentrations than Zn or Cu, and decreased in the order Cr > As > Pb > Cd. No complete Wisconsin dairy ration contained heavy metal concentrations above US maximum acceptable concentrations (NRC, 1980) and would be unlikely to induce any toxic effects in dairy cattle. Levels of Cd were generally present at the lowest concentration of all heavy metals, but appeared to show the greatest long-term risk for exceeding maximum acceptable concentrations (NRC, 1980) in the complete dairy ration if whole farm levels of Cd were to increase through importation of feed or other sources (e.g., agrochemicals and atmospheric deposition).

The main source of Zn, Cu, Cr, As, and Cd in the complete dairy feed ration originated from imported feed (corn-based grain, soybean based-protein mixes, and mineral mixes). The main source of Pb originated from homegrown feeds, possibly through atmospheric deposition of Pb directly onto crops.

The continued importation of heavy metals into dairy feed is likely to be associated with accumulation of these metals in soils where manure is applied. Although the cycling of many heavy metals through the dairy food chain will be limited by factors such as sorption capacity of the soil, pH, salinity, and phytotoxicity of the metal, these may be less limiting for Cd. It is important that sources of Cd in the dairy system are identified and minimized to prevent problems associated with Cd accumulation in the dairy soil system that might arise over the long-term.

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